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pH-Dependent Phase Behaviour of Carbohydrate-based Gemini Surfactants. The Effect of the Length of the Hydrophobic Spacer.

Supplementary Information

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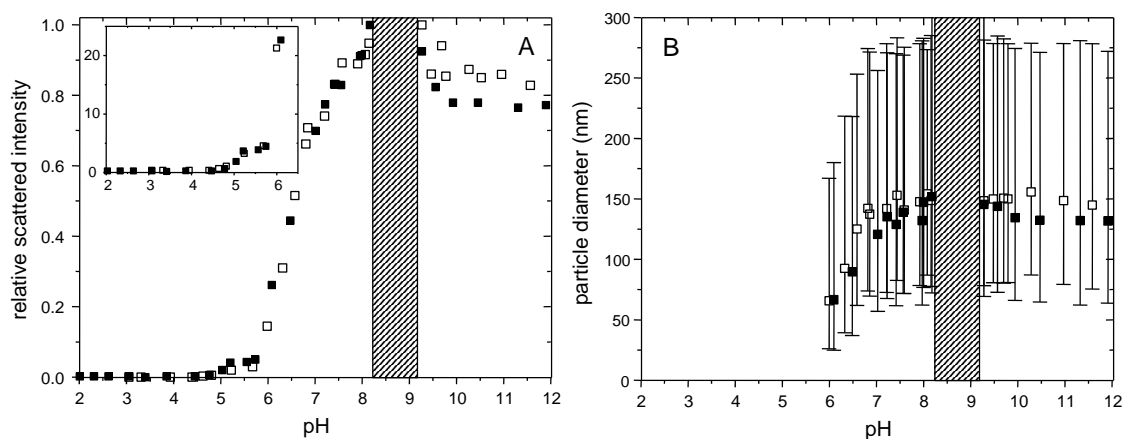


Figure 1. Relative scattered intensity (A) and size distributions (B) of solutions containing gemini amphiphile **3** as a function of pH. The insert in A displays the absolute scattered intensity. Error bars denote the width of the size distribution.¹ Different symbols represent independent experiments starting from stock solutions prepared at near neutral pH. Lined bar denotes the pH region where data could not be obtained due to flocculation.

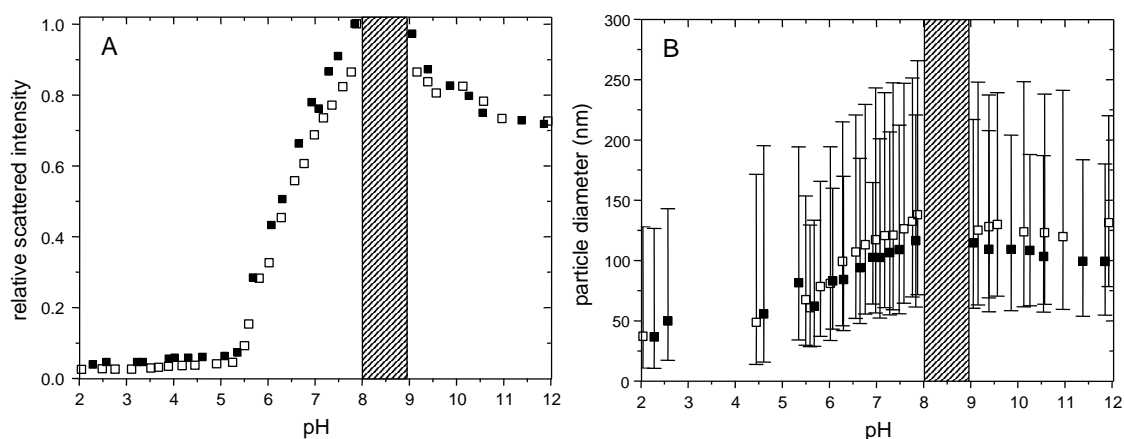


Figure 2. Experimental data for solutions containing gemini amphiphile **2**. For legend details see Figure 1.

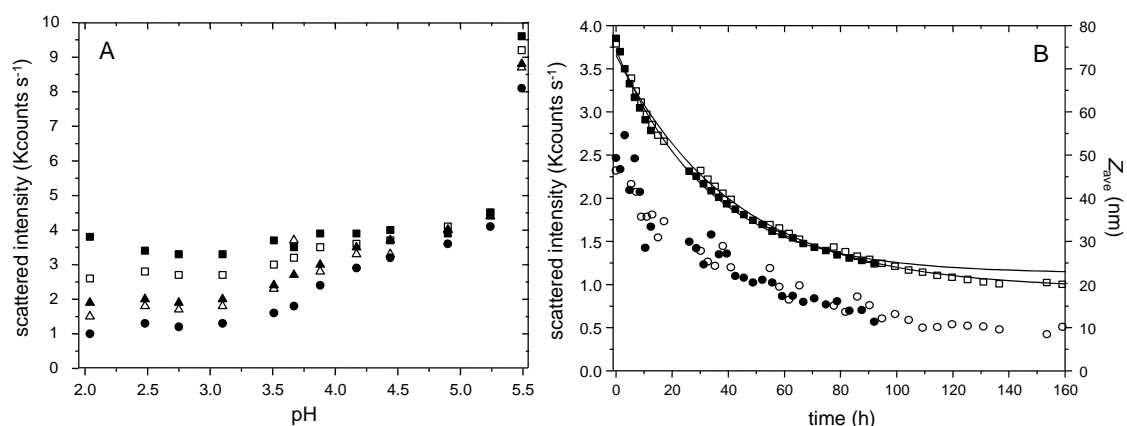


Figure 3. Plots (A) of the scattered intensity (left axis; squares) and the Z_{ave} (right axis; circles) of solutions containing gemini amphiphiles **2** as a function of time after adjusting the pH to 2.3 (solid symbols) and pH 2.0 (open symbols). The lines are exponential fits to the scattered intensity. Note that Z_{ave} can be fitted using the same rate constants (not shown). Plots (B) of the scattered intensity as a function of pH allowing the vesicular solutions to equilibrate for 0 days (■), 1 day (□), 2 days (▲), 3 days (△) and 6 days (●).

Upon allowing solutions containing aggregates formed from **2** to equilibrate with time (Figure 3A), it was noted that above pH 3.7 the relative scattered intensity did not change significantly, whereas below this pH it steadily decreased to about 25% of its original value. At pH 2.0 and 2.3 the intensity of scattered light and the average size of the aggregates (Z_{ave}^2) were recorded in more detail (Figure 3B). It can be seen that the intensity of scattered light and Z_{ave} can nicely be fitted to an exponential equation indicating a slow transformation to more stable structures. This observation might be explained by the observation that after a short equilibration time at low pH a few vesicles are observed in the cryo-electron microscopy pictures that are no longer present after 13 days (Figure 4C).

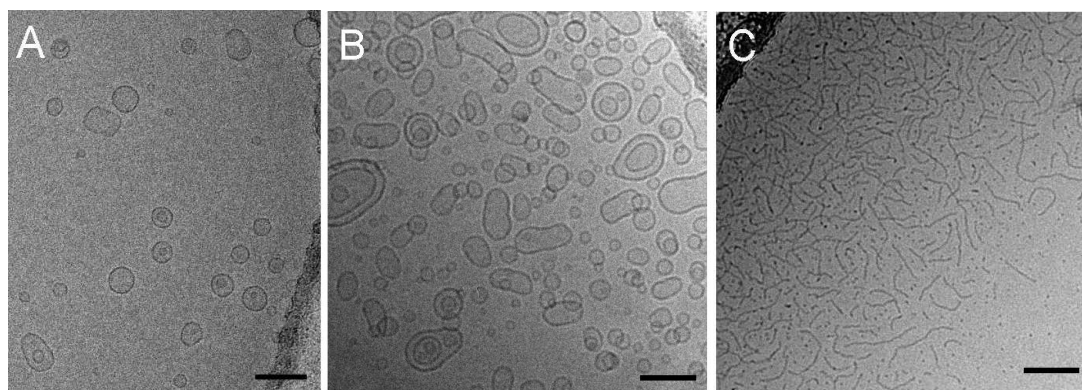


Figure 4. Cryo-electron microscopy pictures of **2** equilibrated over night at pH 7.7 (A), pH 6.4 (B) and pH 2.1 (C; 13 days equilibration). Bar represents 100 nm.

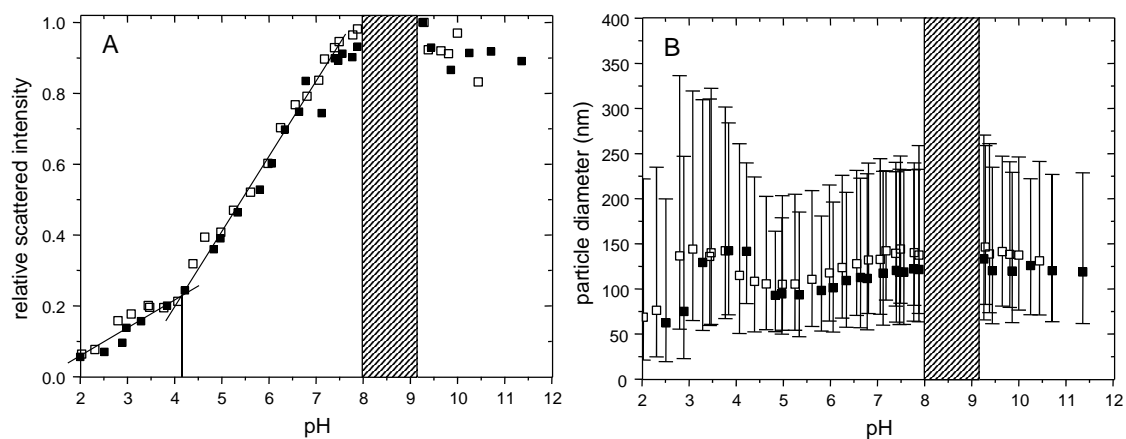


Figure 5. Experimental data for solutions containing gemini amphiphile **1**. For legend details see Figure 1.

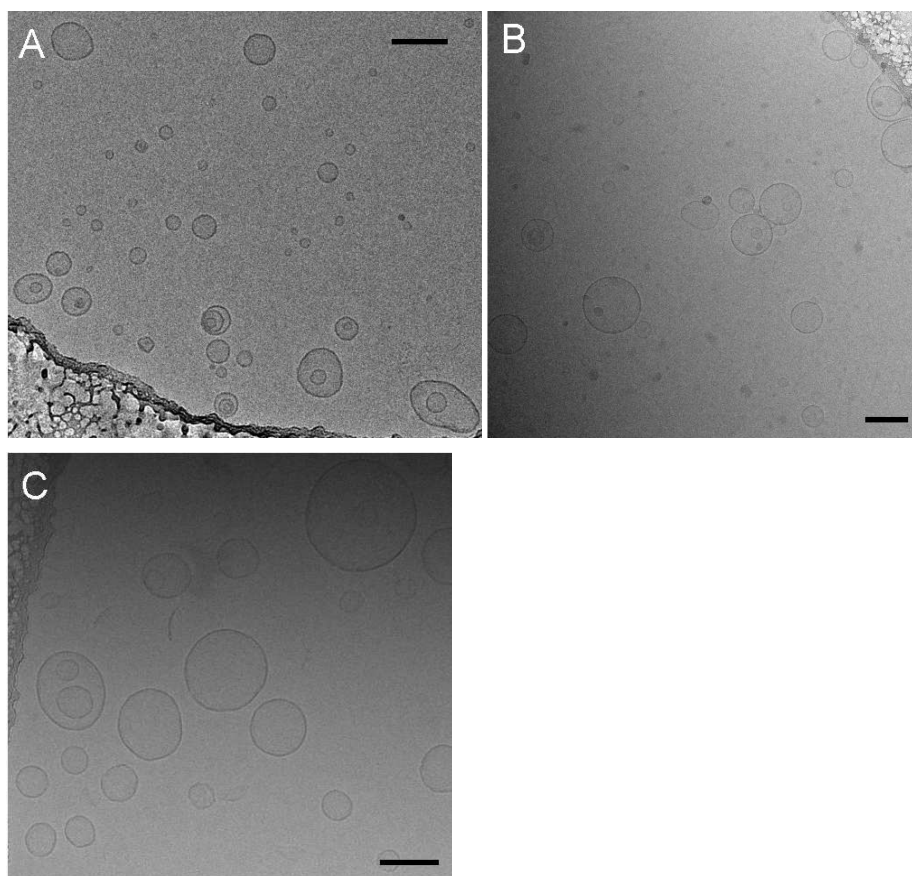


Figure 6. Cryo-electron microscopy pictures of **1** equilibrated over night at pH 10.2 (A), pH 6.7 (B) and pH 4.9 (C). Bar represents 100 nm.

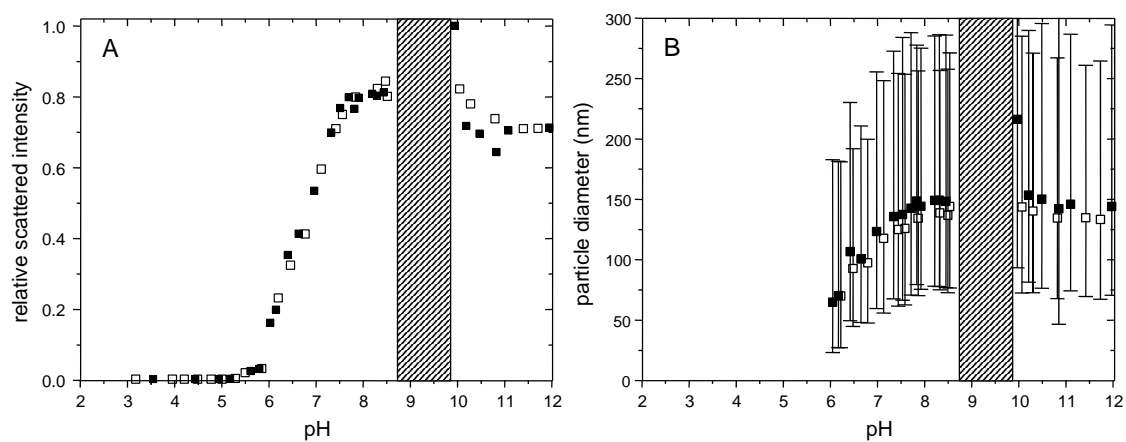


Figure 7. Experimental data for solutions containing gemini amphiphile **4**. For legend details see Figure 1.

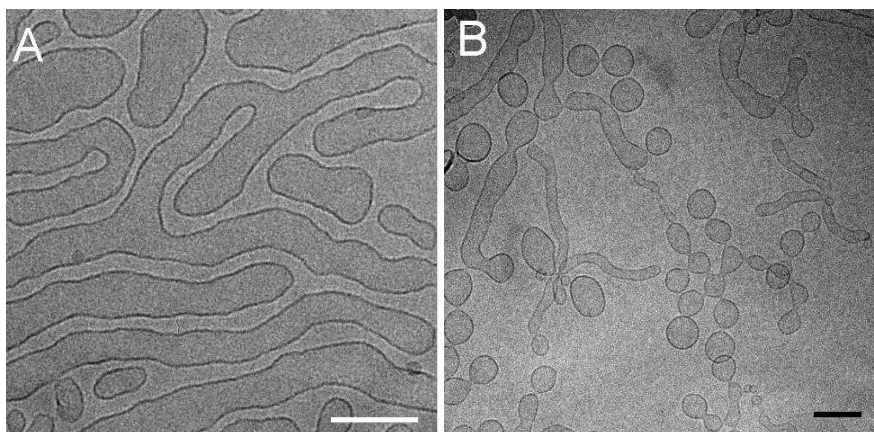


Figure 8. Cryo-electron microscopy pictures of **5** equilibrated over night at pH 7.4 (A) and pH 11.4 (B, supernatant). Bar represents 100 nm.

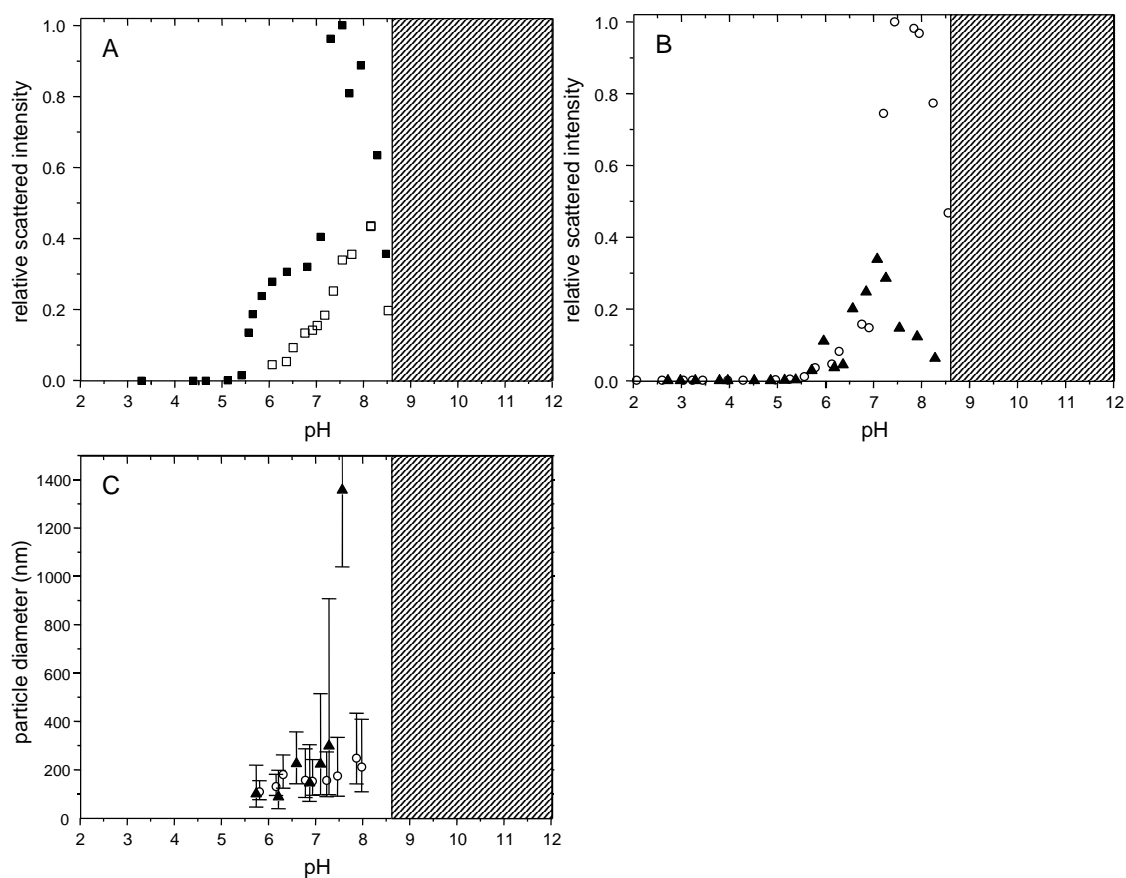


Figure 9. Experimental data for solutions containing **6**. For legend details see Figure 1. Different symbols represent independent experiments starting from stock solutions prepared at near neutral pH (A) or from acidic pH (B,C).

References

- (1) Size distributions were obtained by fitting a Gaussian function to intensity as function of the logarithm of the particle diameter. The width of the size distribution is then the width at half height.
- (2) Z_{ave} is the particle diameter assuming there is no size distribution. Therefore, the value is independent of the used analysis algorithm. Therefore this number gives only an approximation of the average size of the particles. In our case, Z_{ave} is usually close to maximum of the size distribution (data not shown)